# Influence of corrosion of SS316L bipolar plate on PEFC performance

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## HIGHLIGHTS

► A single cell having the SS316L bipolar plate only on the anode side was evaluated.
► The main cause of the performance decline was an increase in contact resistance between the bipolar plate and the GDL.
► The influence of the leaching of Fe ion on the GDL property was negligible.

## ARTICLE INFO

## ABSTRACT

This paper presents an investigation concerning the effect of metallic bipolar plate corrosion on reducing the power generation performance of a polymer electrolyte fuel cell (PEFC). The power generation performance of a single cell having a stainless steel (SS316L) bipolar plate only on the anode side was evaluated experimentally. The results made clear the corrosion behavior on the bipolar plate and the cause of the decline in power generation performance of the cell. The principal cause of the performance decline was an increase in contact resistance between the bipolar plate and the gas diffusion layer (GDL). That is attributed to an increase of iron oxide content in a passive film over the entire bipolar plate surface and precipitation of thick iron oxides in the gas downstream area. The leaching of Fe ions outside the cell was negligible, as was their permeation from the anode to the cathode. Accordingly, there were virtually no signs of an accelerated decline in the conductivity of the electrolyte or its decomposition.

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## 1. Introduction

Changing the previously used carbon bipolar plates to metallic plates has contributed substantially to reducing the size and improving the performance of the fuel cell stack used on fuel cell vehicles (FCVs) [1,2]. On the other hand, in order to reduce the cost of the fuel cell stack further, it is desirable to adopt a low-cost base metal for metallic bipolar plates and an inexpensive surface treatment process. Commercial grade stainless steel in particular is regarded as a promising material for the base metal.

However, as is generally known, metallic bipolar plates corrode in the severe environment inside a fuel cell, and such corrosion may reduce the power generation performance of a cell. In this regard, electrochemical evaluations have been conducted using commercial grade stainless steel [3–12]. It has also been reported that metal ions leached from metallic bipolar plates reduce the conductivity of

the electrolyte [13,14] and that OH radicals produced from a reaction with hydrogen peroxide are related to decomposition of the electrolyte [15–17]. However, many of these reports concern evaluations conducted using a three-electrode electrochemical test or evaluations with the test piece of electrolyte membrane under conditions simulating those of the fuel cell environment. There are few reports of studies that examined metallic bipolar plate corrosion or conductivity in actual fuel cell evaluations or that investigated in detail the effect of such corrosion on other fuel cell components. Therefore, in this study power generation tests of a fuel cell with a metallic bipolar plate were conducted to investigate bipolar plate corrosion behavior and resultant changes in power generation performance for the purpose of confirming the consistency with the findings reported so far.

Evaluations were carried out with a bipolar plate made of SS316L as the base metal that was not surface-treated (denoted as 316L bipolar plate). Evaluating cells in which various base metals are used to make the bipolar plate placed at one electrode side makes it possible to confirm the effect of the corrosion behavior of the bipolar plate at each electrode side as well as the impact on power generation performance. Therefore, a single test cell was

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prepared in which the 316L bipolar plate was positioned on the anode side and a carbon bipolar plate was used on the cathode side. There were two reasons for adopting this configuration.

1) To confirm the corrosion behavior of bipolar plate on the anode side and its effect on power generation performance.
2) To confirm the effect of leached metal ions on decomposition of the electrolyte membrane, as the anode side is reported to be more sensitive in this regard [17].

## 2. Experimental

### 2.1. Single cell preparation

A PRIMEA® Series 5570 membrane electrode assembly (MEA, W.L. Gore & Associates, Co., Ltd.) was used in this study. This MEA integrated a 30-μm-thick perfluorosulfonic acid membrane and the catalyst layer having a Pt loading of 0.4 mg cm$^{-2}$ and was sandwiched on both sides by a 190-μm-thick gas diffusion layer (GDL) made of carbon paper. The active area of was 25 cm$^2$.

The metallic bipolar plate was made of 0.1-mm-thick 316L on which 23 straight flow channels were stamped. The flow channel width and the rib width were both 1 mm and the depth was 0.5 mm. The 316L bipolar plate was positioned on the anode side to investigate its corrosion behavior and the effect on power generation performance of metal ions leached from the plate. A carbon bipolar plate assumed to be free of material degradation and ion leaching was used on the cathode side. The cell with the 316L bipolar plate on the anode side will be denoted as the 316L cell in the following discussion. Only the rib surface of the 316L bipolar plate was polished with #1200 abrasive paper, after which it was cleaned in an acetone and deionized water.

In the single cell assembly process, a pressure-sensitive film (Prescale, Product Code LLW, Fuji Film Co., Ltd.) [18] was inserted between the bipolar plate and the GDL in advance. The cell component was then compressed by applying surface pressure of 2 MPa to the bipolar plate rib surface.

### 2.2. Cell operation and diagnostic tests

The MEA was conditioned before conducting a cell performance evaluation. Conditioning was performed by applying a current load cycle between the open circuit voltage (OCV) and a current density of 1.0 A cm$^{-2}$ and further by holding at 0.8 A cm$^{-2}$ for 12 h. The conditions used were a cell temperature of 70 °C, atmospheric operating pressure, H$_2$ supply to the anode at a flow rate of 0.5 dm$^3$ min$^{-1}$, air supply to the cathode at a flow rate of 1.0 dm$^3$ min$^{-1}$ and humidification of both gas streams to 100% RH.

To avoid the corrosion of the 316L bipolar plate, conditioning was conducted with carbon bipolar plates positioned at both anode and cathode sides (The cell assembled with carbon bipolar plates at both sides will be denoted as the carbon cell in the following discussion.).

Power generation tests were conducted for 300 h at a load current of 0.008 A cm$^{-2}$ under conditions of a cell temperature of 70 °C, H$_2$ supply to the anode at a flow rate of 0.5 dm$^3$ min$^{-1}$, air supply to the cathode at a flow rate of 1.0 dm$^3$ min$^{-1}$ and with both gas humidified to 60%RH. There are reports about the degradation behavior of the catalyst layer and the electrolyte membrane during long term tests with a single cell [19,20]. Such degradation was evaluated under an OCV condition because the degradation occurred under a high potential state. In this study, a condition of vehicle idling was assumed and a low current load was applied as noted above. This condition is effective for the purpose of evaluating bipolar plate corrosion and its effect on power generation performance with respect to the two points below.

i) High cell voltage provides a severe condition for evaluating the durability of the electrolyte membrane, so there is a possibility that fluoride ions and sulfuric acid ions originating from decomposition of the electrolyte might promote corrosion of the metallic bipolar plate.
ii) The process of generating power produces protons and attendant osmotic drag water in the electrolyte membrane [21]. When metal ions leached from the metallic bipolar plate pass through the electrolyte membrane together with the accompanying water, they might promote degradation of the electrolyte.

The current–voltage (IV) characteristic, which is an index of the power generation performance of a cell, was measured after switching the O$_2$ supply to the cathode to a flow rate of 2.0 dm$^3$ min$^{-1}$ (60%RH) in order to eliminate oxygen diffusion overvoltage on the cathode side. The carbon cell was also evaluated under the same conditions in order to compare with the 316L cell. The Cyclic voltammetry (CV) and electrochemical impedance spectroscopy (EIS) were conducted under a flow rate of 0.2 dm$^3$ min$^{-1}$ for both the N$_2$ and the H$_2$ supplied to the anode and cathode, respectively.

Drained water discharged from the cell during the 300 h power generation test was collected by cooling the exhaust gas pipe to 5 °C. Qualitative and quantitative analyses for the leaching ions were conducted by ion chromatography and inductively coupled plasma mass spectrometry (ICP-MS).

### 2.3. Measurement of contact resistance

The experimental setup used in measuring the contact resistance between the GDL and the bipolar plate is shown in Fig. 1. Electrical resistance was measured using a four-terminal configuration. The bipolar plate was sandwiched between two GDLs and compressed by copper electrodes 20 mm in diameter; a direct current of 1 A was applied and the voltage drop between the terminals was measured. The voltage was measured at a voltage terminal 1 mm in diameter that was positioned in the center of the copper electrodes and was
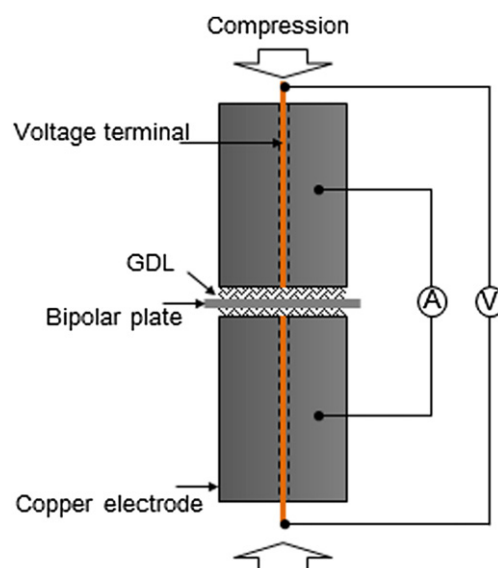


**Fig. 1.** Schematic configuration of electrical resistance measurement setup.
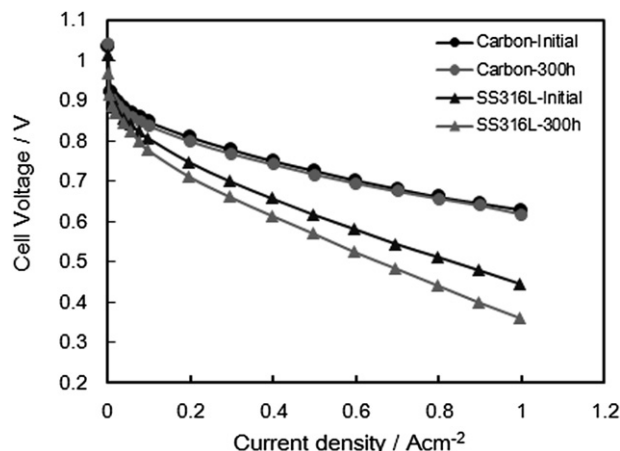
**Fig. 2.** Changes in IV performance during 300 h test. Carbon: carbon cell, SS316L: 316L cell.



**Fig. 4.** Changes in EIS during 300 h test.

insulated from them. Based on the electrical resistance obtained from the relation $R = V/I$, contact resistance ($R_{CR}$) was calculated as

$$R_{CR} = S_1(R_{all} - 2R_{GDL})/2 \qquad (1)$$

where $R_{all}$ denotes all the measured resistance, $R_{GDL}$ is the through-plane resistance of the GDL, which was measured for one GDL sandwiched between the copper electrodes, and $S_1$ indicates the area of the copper electrodes [22].

### 2.4. Measurement of GDL degradation

The effect of the metal ions leached from the metallic bipolar plate on the electrical and mechanical properties of the GDL was investigated. Only Fe ions were considered as metal ions in this work. Aqueous solutions consisting of 20 and 100 ppm of $Fe(NH_4)_2(SO_4)_2 \cdot 6H_2O$ were prepared. A GDL (TGP-H-60, Toray Industries Inc. [23]) measuring $30 \times 30$ mm was immersed in a 70 ml solution at 80 °C for 300 h and changes in its electrical and mechanical properties were measured with the contact resistance measurement setup shown in Fig. 1.

## 3. Results and discussion

### 3.1. Cell performance and diagnostic test

Fig. 2 shows the IV characteristics measured for the 316L cell before and after the 300 h test in comparison with the carbon cell.
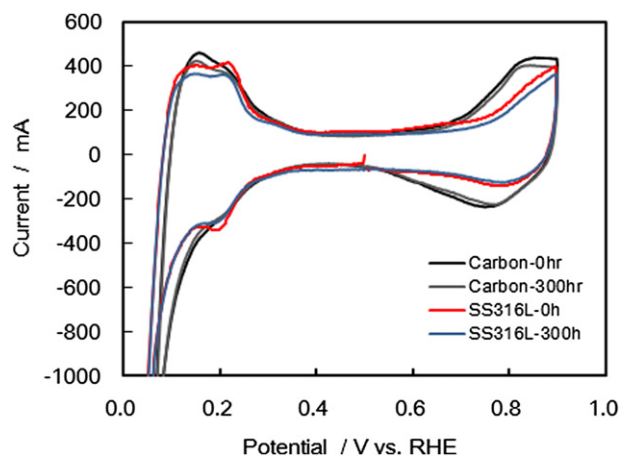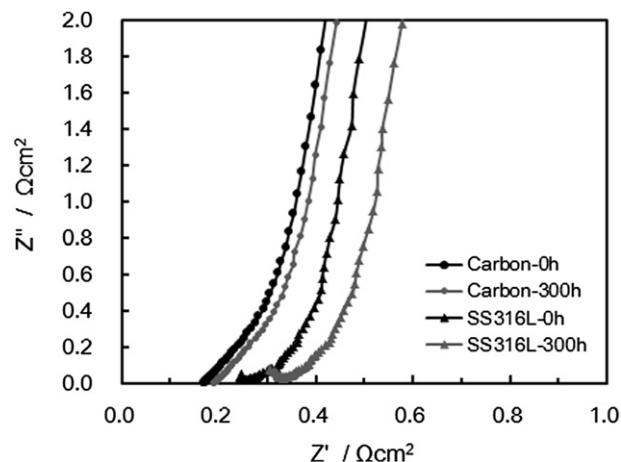
After 300 h, the IV characteristic of the carbon cell showed a decline in cell voltage of 11 mV at a current density of 1 A cm$^{-2}$. Assuming that carbon bipolar plates do not deteriorate chemically, this voltage drop is attributed to the degradation of the MEA that occurred with elapsed time under the conditions of the test. It is seen that the initial cell voltage of the 316L cell was markedly lower than that of the carbon cell. Presumably, the contact resistance between the bipolar plate and the GDL contributed
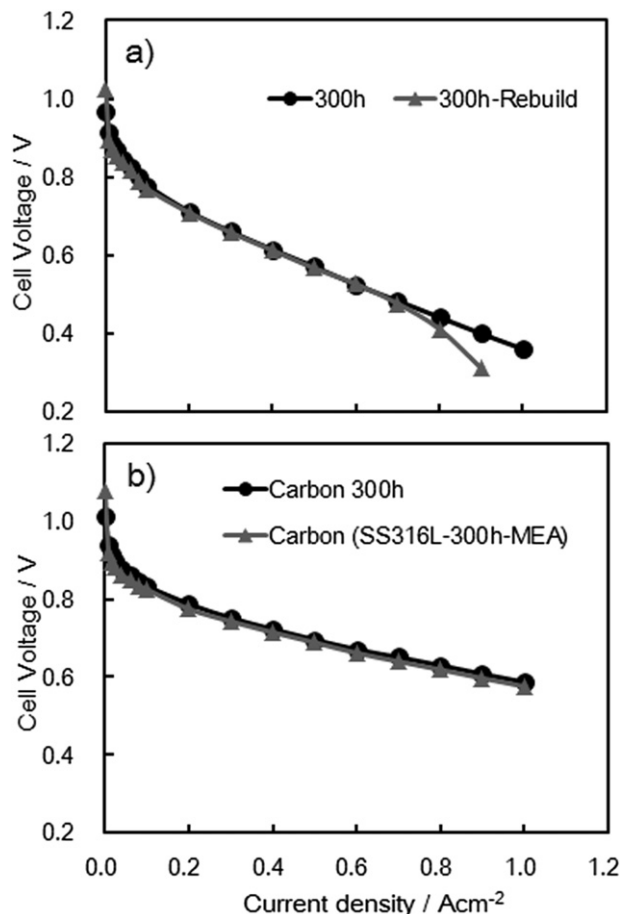


**Fig. 3.** Changes in CV during 300 h test.



**Fig. 5.** Changes in IV performance: a) rebuilt after 300 h test, b) carbon cell using the MEA employed in the 316L cell during 300 h test.

significantly to this difference in the initial IV characteristic. Following the 300 h test, it was found that the 316L cell showed a voltage drop of 85 mV (at 1 A cm$^{-2}$) from the initial voltage.

The CV and EIS measured results are shown in Figs. 3 and 4, respectively. The electrochemical area (ECA) calculated from the CV results showed a decline of less than 3% for the 316L cell, indicating that there was very little possibility of it affecting power generation performance. Additionally, the change in the electrolyte resistance calculated from the high frequency in the EIS correlated well with the IV characteristic. Fig. 5-a shows the IV characteristics measured for the 316L cell that was rebuilt after the 300 h test by simply disassembling and reassembling of the same cell. If the corrosion products precipitated on the bipolar plate surface during the 300 h test, the electrical paths between the bipolar plate and the GDL were fixed and maintained with them. In that case, the cell voltage decreased drastically after the reassembling of the cell because the electrical paths were disconnected. However, although the cell voltage decreased at current densities above 0.8 A cm$^{-2}$, the decline in cell voltage due to the resistance overvoltage was steady between current densities of OCV and 0.7 A cm$^{-2}$. This result inferred

that there were no corrosion products and large voltage drop of 85 mV after the test attributed to the change of the passive film property on 316L plate.

On the other hand, using the MEA employed in the 316L cell during the 300 h test, the IV characteristics were measured with the carbon cell. The results are shown in Fig. 5-b, where the IV characteristics of the carbon cell in the 300 h test are also shown for comparison. The 316L cell incorporating the used MEA showed a decline in cell voltage of 11 mV (at 1 A cm$^{-2}$) compared with the carbon cell. While a slight decline in cell performance was seen, no pronounced change in performance due to the metallic bipolar plate was observed.

## 3.2. Corrosion and surface profile

### 3.2.1. Surface morphology

The corrosion on the bipolar plate surface was observed on the ribs in the gas downstream area and around the gas outlet manifold. Fig. 6 shows SEM photographs of corrosion products in the gas downstream area. In this area reddish-brown oxide precipitates
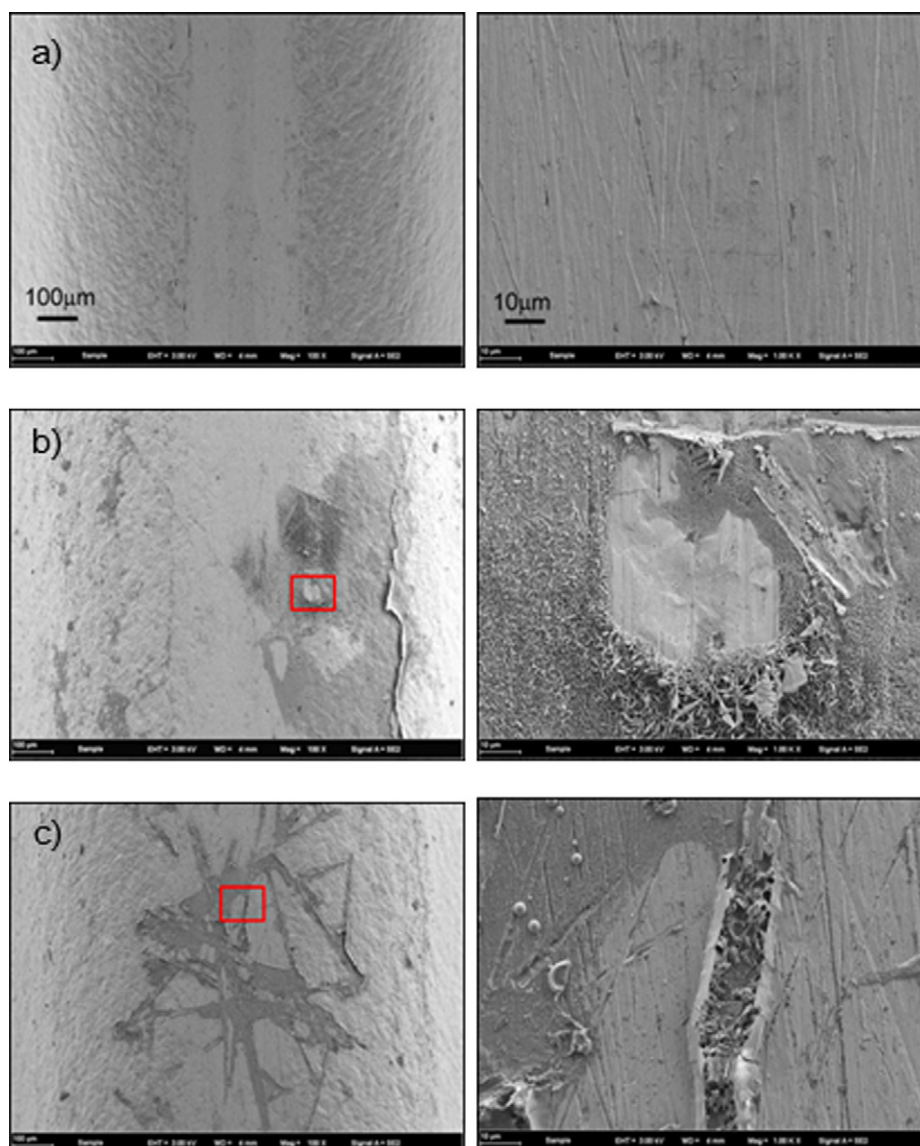


Fig. 6. SEM images of SS316L bipolar plate on the rib in the gas downstream area: a) initial (as polished), b) and c) after 300 h test.
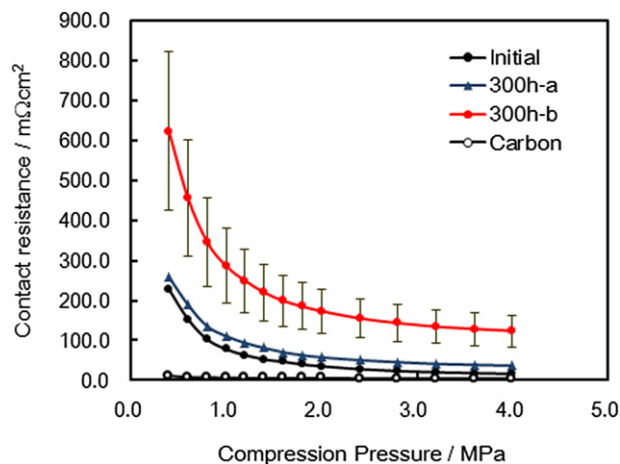
**Fig. 7.** Contact resistance between bipolar plate and GDL under various compression pressures: 300 h-a) at the center of the 316L bipolar plate after 300 h test, 300 h-b) in the gas downstream area of 316L bipolar plate after 300 h test.

were observed (Fig. 6-b). The crystal shapes of the precipitates and results of an SEM–EDX analysis revealed that they were iron oxides. In addition, line-like corrosion was also observed at multiple locations on the ribs of the bipolar plate (Fig. 6-c) in the same area. The width of the line-like corrosion was approximately 10 μm, roughly the same size as the width (8 μm) of the carbon fibers. This implies that iron oxides accumulated around the carbon fibers in contact with the bipolar plate. Spherical-shaped oxides presumed to be water droplet marks were also observed (Fig. 6-c), suggesting that liquid water which remained in the gas downstream area affected bipolar plate corrosion.

### 3.2.2. Interfacial contact resistance

Fig. 7 shows the contact resistance found between the 316L bipolar plate and the GDL in comparison with that measured between the carbon bipolar plate and the GDL. The initial contact resistance of the 316L bipolar plate before the 300 h test was 35.5 mΩ cm$^2$ at a compression pressure of 4 MPa, which was higher than the corresponding value of 5.0 mΩ cm$^2$ for the carbon bipolar plate at the same compression pressure. After the 300 h test, the contact resistance of the bipolar plate along the in-plane gas flow direction increased to 57.7 mΩ cm$^2$ in the center section from the gas inlet upstream and to 174 mΩ cm$^2$ in the gas downstream area (both values for a compression pressure of 4 MPa, denoted as 300 h-a and 300 h-b in Fig. 7 respectively). Accordingly, it is assumed that the voltage drop of 85 mV (at 1 A cm$^{-2}$) observed in the IV characteristic of the 316L cell was caused almost entirely by the combined increase in contact resistance in the center section and in the gas downstream area. In actuality, the in-plane electrical resistance of the GDL adjacent to the bipolar plate is assumed to be very small. Therefore, the electric current does not flow in the corroded regions of the bipolar plate that have high contact resistance. Instead, the electric current also flows along the in-plane direction of the GDL toward areas of low contact resistance. In such cases, the electric current concentrates in uncorroded areas of the bipolar plate, which probably increases the IR-drop.

### 3.2.3. AES profiles

After the 300 h test, an auger electron spectroscopy (AES) surface analysis was performed near the center of the bipolar plate where there were no apparent corrosion and in the gas downstream area where corrosion was observed. The thickness of the oxide film was 3.3 nm before the test (Fig. 8-a) and 4.2 nm near the center of the bipolar plate after the test (Fig. 8-b), indicating that
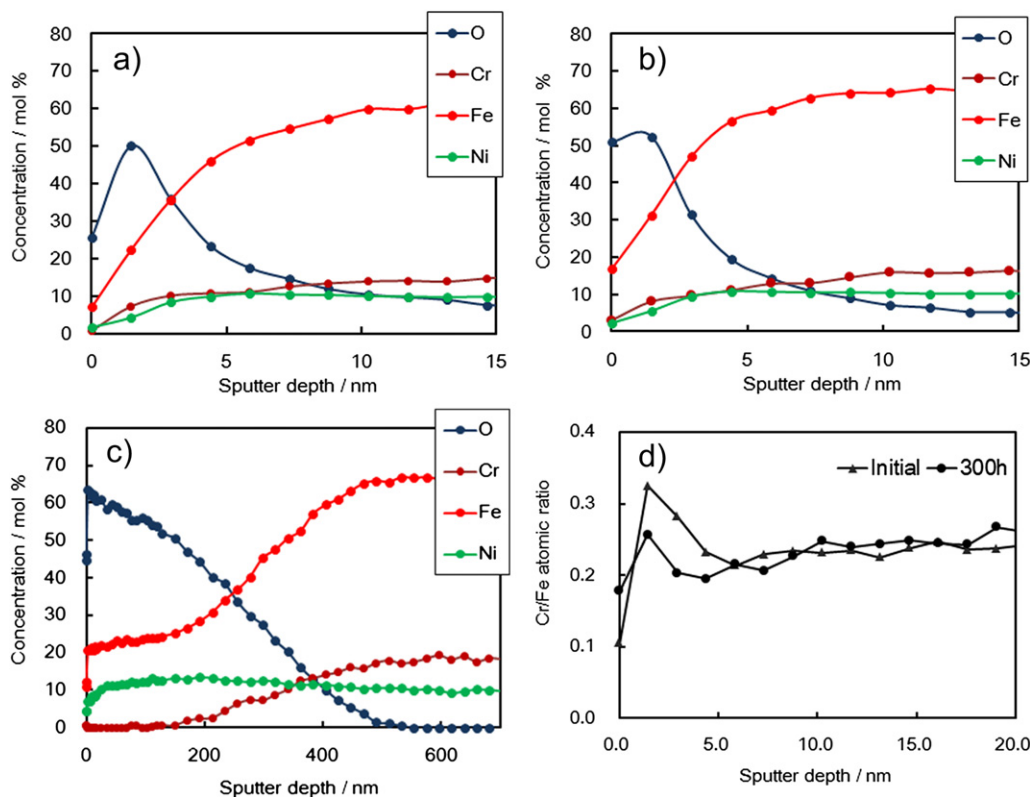


**Fig. 8.** AES depth profiles of passive film on the rib of the 316L bipolar plate: (a) initial (as polished), (b) at the center of active area after 300 h test, (c) in the gas downstream of active area after 300 h test, and (d) Cr/Fe atomic ratio.
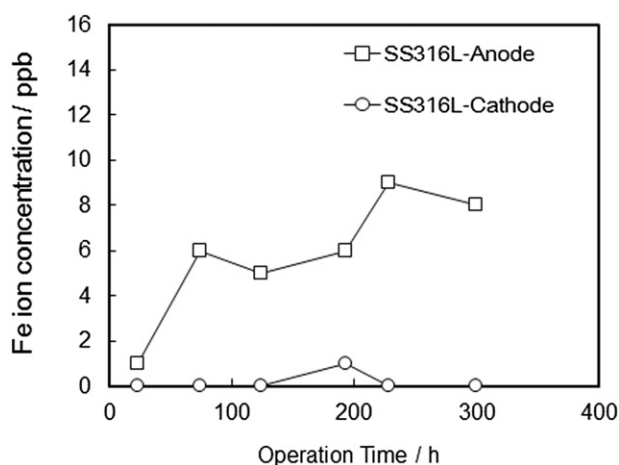
**Fig. 9.** Changes in $Fe^{2+}$ emission in the drain water from the 316L cell.

the oxide film thickness was virtually unchanged. In the gas downstream area (Fig. 8-c) where the iron oxide precipitates were observed, the thickness was found to be markedly larger at 270 nm.

The change in the Cr/Fe atomic ratio calculated from Fig. 8-a and -b is shown in Fig. 8-d. The ratio of the iron oxide content from the surface to a depth of 8 nm increased even in the center of the bipolar plate where no corrosion was observed. It can be inferred that the iron oxides in the Cr passive film increased the electrical resistance of the bipolar plate surface, thereby increasing the contact resistance with the GDL. Several investigations of the Cr/Fe atomic ratio of bipolar plate metals have been reported. Kumagai et al. [6] reported that the contact resistance between the GDL and a SS310S bipolar plate was higher when polarization took place in an aqueous solution of pH 5.5

than in that of pH 1.2. They also noted that an oxide layer with an atomic ratio centered on $Cr^{3+}$ ions formed on the surface of as-polished bipolar plates and on plates polarized in aqueous solutions of pH < 3.3, whereas the proportion of $Fe^{3+}$ ions was dominant in oxide layers formed on bipolar plates polarized at pH 5.5. Feng et al. [7] investigated the pH dependence of the contact resistance and the Cr/Fe atomic ratio for SS316L bipolar plates. They conducted polarization tests with the three-electrode method in aqueous solutions of pH 3–6 that simulated the anode environment inside the cell and reported an increase in the Cr concentration on the bipolar plate surface. They attributed that increase to the selective dissolution of Fe ions from the bipolar plate surface. Diffusion of Fe ions into an aqueous solution can easily take place in polarization tests, but Fe ions did not readily diffuse from the bipolar plate surface (i.e., be leached outside the cell) during the cell evaluations conducted in this study. Therefore, it is assumed that they remained on the bipolar plate surface as oxides.

### 3.3. Ion leaching and acidity of drained water

Fig. 9 shows the change in the Fe ion concentration in the drained water discharged from the cell during the 300 h test. The concentration of Fe ions was higher on the anode side because the 316L bipolar plate was positioned there. Fe ions detected on the cathode side are usually leached from the anode side and permeate through the MEA, but virtually no Fe ions were detected on the cathode side in this test. Taking into account the SEM photographs in Fig. 6, it is assumed that larger quantities of Fe ions leached from the bipolar plate precipitated as oxides on the plate surface or around the carbon fibers than were discharged from the cell as ions. Ma et al. [13] investigated the effect of the Fe ion concentration on reducing electrolyte conductivity and reported that the presence of 10 ppm of Fe and Cr ions sharply reduced the through-plane conductivity of a Nafion 112 membrane. Wang et al. [14] also reported that the
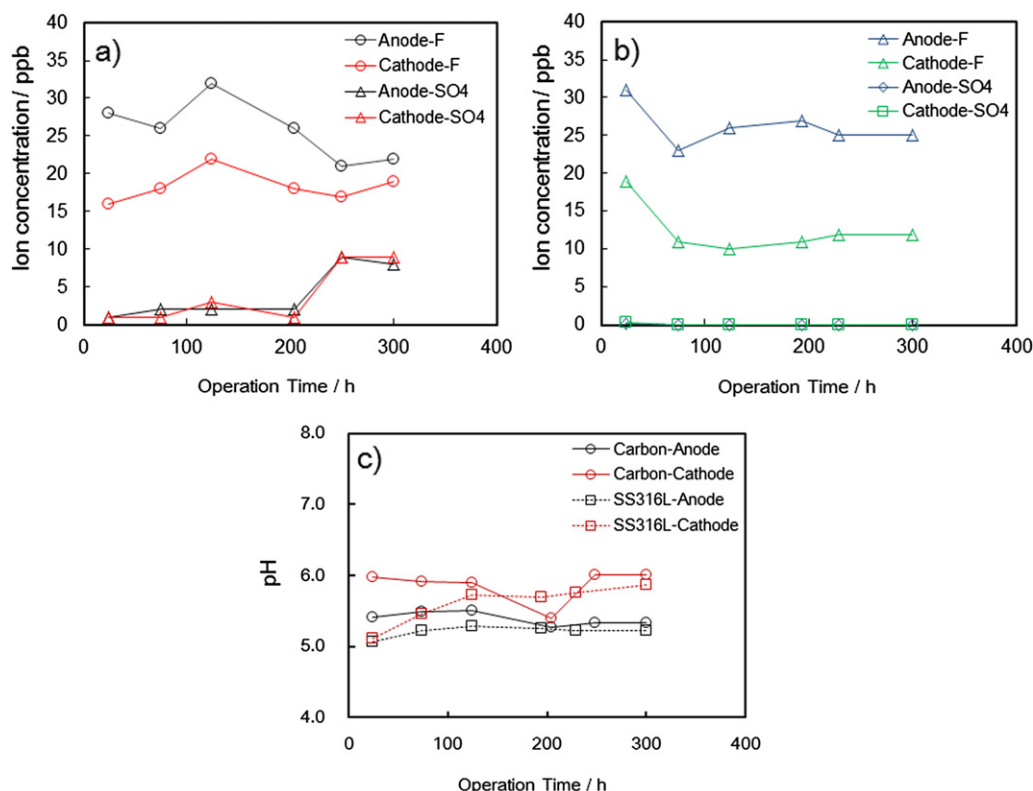


**Fig. 10.** Changes in $F^-$ and $SO_4^{2-}$ ion concentration and the pH value of the drain water: a) 316L cell, b) carbon cell, c) pH.

in-plane conductivity of Nafion membrane decreased in the presence of Fe ion concentration of 300 ppm or more. Taking those results into consideration, it is assumed that the conductivity of the electrolyte showed virtually no decline in this test because the quantity of Fe ions was very small.

Fig. 10-a shows the change in the $F^-$ and $SO_4^{2-}$ ion concentrations in the drained water. For the sake of comparison, the evaluation results are shown in Fig. 10-b for the carbon cell. The results indicate that the quantity of $F^-$ ions did not differ significantly between the carbon cell and the 316L cell. The quantity of $SO_4^{2-}$ ions was markedly small and less than the quantity of $F^-$ ions. The quantity of electrolyte that decomposed during 300 h was calculated from the quantity of $SO_4^{2-}$ ions and the specific gravity and ion exchange capacity of the electrolyte; it was found to be 0.3% of the total electrolyte weight. At this time, however, the decomposed position of electrolyte is not clear. Moreover, the pH value of the drained water was correlated with the quantity of ions and no appreciable difference was found between the carbon cell and the 316L cell (Fig. 10-c). The foregoing results imply that the quantity of Fe ions, which would cause corrosion of the 316L bipolar plate, was markedly small and also that the electrolyte did not undergo any accelerated decomposition.

### 3.4. Influence of Fe ions on electrical and mechanical properties of GDL

Two factors can be considered that might cause changes in the electrical properties of the GDL due to the leaching of Fe ions from metallic bipolar plates. One is an increase in the contact resistance between the carbon fibers composing the GDL because of metal oxides adhering to the fiber surface. The other factor is an increase in contact resistance between the bipolar plate and the GDL due to a change of GDL elasticity because of the same cause. It is known that the water content of the electrolyte in fuel cells used on FCVs changes according to the power generation conditions, resulting in expansion and contraction that vary the compressive pressure on fuel cell components. The adhering of metal oxides to the carbon fibers in particular can be expected to increase the contact resistance sharply because once the GDL have been compressed they do not show elastic deformation, so the number of contact points with the bipolar plate decreases in relation to a change from high to low compressive pressure.

In this study, electrical resistance was measured as the compressive pressure was reduced from 1.0 MPa. Fig. 11 shows the electrical resistance measured for the GDL that were immersed in aqueous solutions with an $Fe^{2+}$ ion concentration ranging from 20
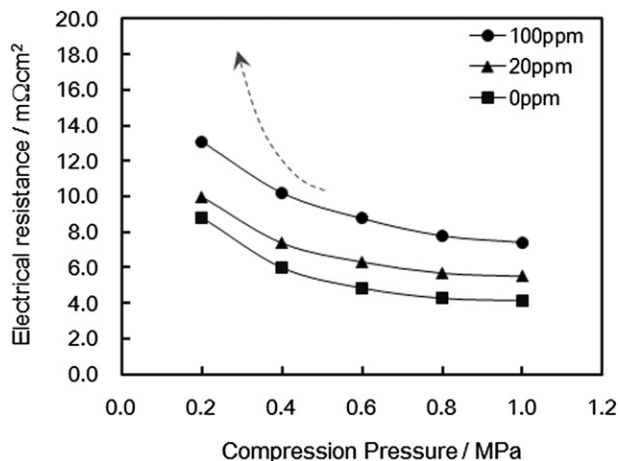


**Fig. 11.** Through-plane electrical resistance of an immersed GDL under a decreasing load condition.
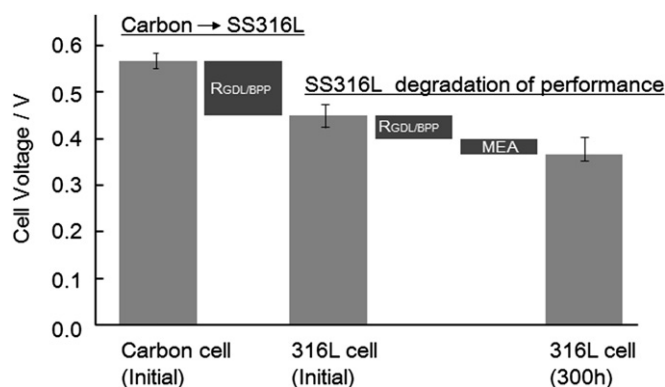


**Fig. 12.** Breakdown of the degradation of the 316L cell performance. $R_{GDL/BPP}$: voltage drop caused by the contact resistance between the GDL and the bipolar plate, MEA: voltage drop caused by MEA degradation.

to 100 ppm. For an aqueous solution without any Fe ions (0 ppm), electrical resistance of 4.2 m$\Omega$ cm$^2$ was measured under compressive pressure of 1 MPa. The resistance measured for the GDL immersed in the 20 ppm and 100 ppm aqueous solutions was 5.5 and 7.4 m$\Omega$ cm$^2$, respectively. Based on these results and the leached Fe ion concentrations shown in Fig. 9 (Fe ion concentration shown at the ppb level), it is inferred that the electrical resistance of the GDL did not increase significantly.

It is also possible that Fe oxides might precipitate on the surface of the carbon fibers inside the GDL, causing the fiber elasticity to decline with the result that the electrical contact points between the fibers would decrease in relation to the reduction of the surface pressure from 1 MPa. In that case, it is estimated that the electrical resistance of the GDL would increase relative to the decline in compressive pressure, as indicated by the dashed line in Fig. 11. However, the results show that none of the samples tested showed any significant change in electrical properties relative to the compressive pressure applied to the GDL.

These results imply that the electrical properties of the GDL did not change significantly due to the use of the 316L bipolar plate or the leaching of Fe ions from the plate.

### 3.5. Breakdown of cell performance degradation

Based on the test results, the factors causing the degradation of the 316L cell generation performance decline were identified and are summarized in Fig. 12. The major factor was an increase in contact resistance between the 316L bipolar plate and the GDL. The degradation of MEA performance in the 300 h test was estimated at a minimum to be a drop of 11 mV observed for the carbon cell (Fig. 2) and a drop of 11 mV observed for the MEA used in the 316L cell (Fig. 5). The degradation of cell generation performance related to the decline in GDL conductivity due to the leaching of Fe ions was no more than 1 mV.

### 4. Conclusions

This study was made of the effect of bipolar plate corrosion on the power generation performance of a single fuel cell when a SS316L bipolar plate was positioned on the anode side. The results of the 300 h test revealed that nearly all of the degradation of cell performance was caused by increased contact resistance between the bipolar plate and the GDL. In the severe environment of anode side, an increase of iron oxide content in a passive film over the entire bipolar plate surface and precipitates of iron oxide in the gas downstream area, thereby increasing the contact resistance.

It was found that Fe ions had very little effect on the electrolyte and GDL. Accordingly, the first prerequisite condition for using SS316L bipolar plates is to provide a surface treatment that maintains low contact resistance. Effective measures in this regard include a surface treatment process that suppresses the selective dissolution of Fe or modification of the composition of the base metal.

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